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SQUEEZE FLOW AND COMPACTION BEHAVIOR OF TOUGHENED POLYIMIDE MATRIX COMPOSITES

by

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As a means of increasing the resistance of fiber composites against *microcracking* or *matrix damage accumulation*, past efforts of toughening brittle thermoset resins with the inclusion of ductile secondary domain or the reduction of crosslink density were successful to various extents. For example, the use of elastomer-toughened epoxy resin matrix became a standard practice of composite prepreg industry. In the case of brittle thermoset polyimide matrix composites, several promising approaches such as the increase of molecular weight between crosslinks (Mc) or the formation of semi-interpenetrating network with thermoplastic polyimide were shown to toughen the matrix very effectively. However, their effectiveness has been limited by the lack of resin flow, the difficulty of removing solvent or other processing difficulties.

In view of the above-discussed limitations, a new research program on the processing science of toughened polyimide matrix composites has been proposed with the 1991 Summer Faculty Research Project (SFRP) as a preliminary phase. The main objectives of the program are: (a) to determine optimum processing conditions for toughened polyimide matrix composites in compression molding as well as filament winding based on the understanding of *squeeze flow, kinetics of cure, kinetics of volatile removal*, and *compaction behavior* of material elements, and (b) to define the roles and interaction of the degree of compaction (void/resin content) and residual stresses at the fiber-matrix interface in controlling the microcrack resistance of toughened polyimide matrix composites.

In the 1991 SFRP, main emphasis was placed upon the squeeze flow and compaction behavior of LaRC-RP series polyimide matrix composites. The composite prepregs were prepared with unsized IM7 graphite fibers and the following matrix resins of varied Mc (and therefore of varied fracture toughness):

	Mc
RP48	1 100
RP46	1 500
RP52	* 2:1 IPN of RP48/RP49
RP50	9 950
RP51	* 1:1 IPN of RP48/RP49
RP47	15 000
RP49	21 000

The measurement of squeeze flow behavior was performed by a plastometer which monitors the change of thickness of a prepreg specimen laid between two parallel plates under the specified temperature and pressure history. A critical evaluation of the plastometer data was attempted by examining the morphology of the specimen at various points during the squeeze flow. The effects of Mc of resin, imidization (B-staging) condition and pressure on the squeeze flow behavior were examined. The following facts were learned:

(1) The plastometer data under static loading provide three key results: melt transition temperature of linear polyimide resins, the degree of resin squeeze flow, and processing window.

(2) In estimating the degree of squeeze flow, an initial specimen thickness has to be defined <u>before</u> B-staging or imidization. The imidization step results in the increase of prepreg thickness because of resin foaming and the reduction of prepreg width. The use of the prepreg thickness after imidization as a baseline leads to serious errors in the estimation of the degree of flow. Higher prepreg thickness after imidization is observed with increasing Mc of resins.

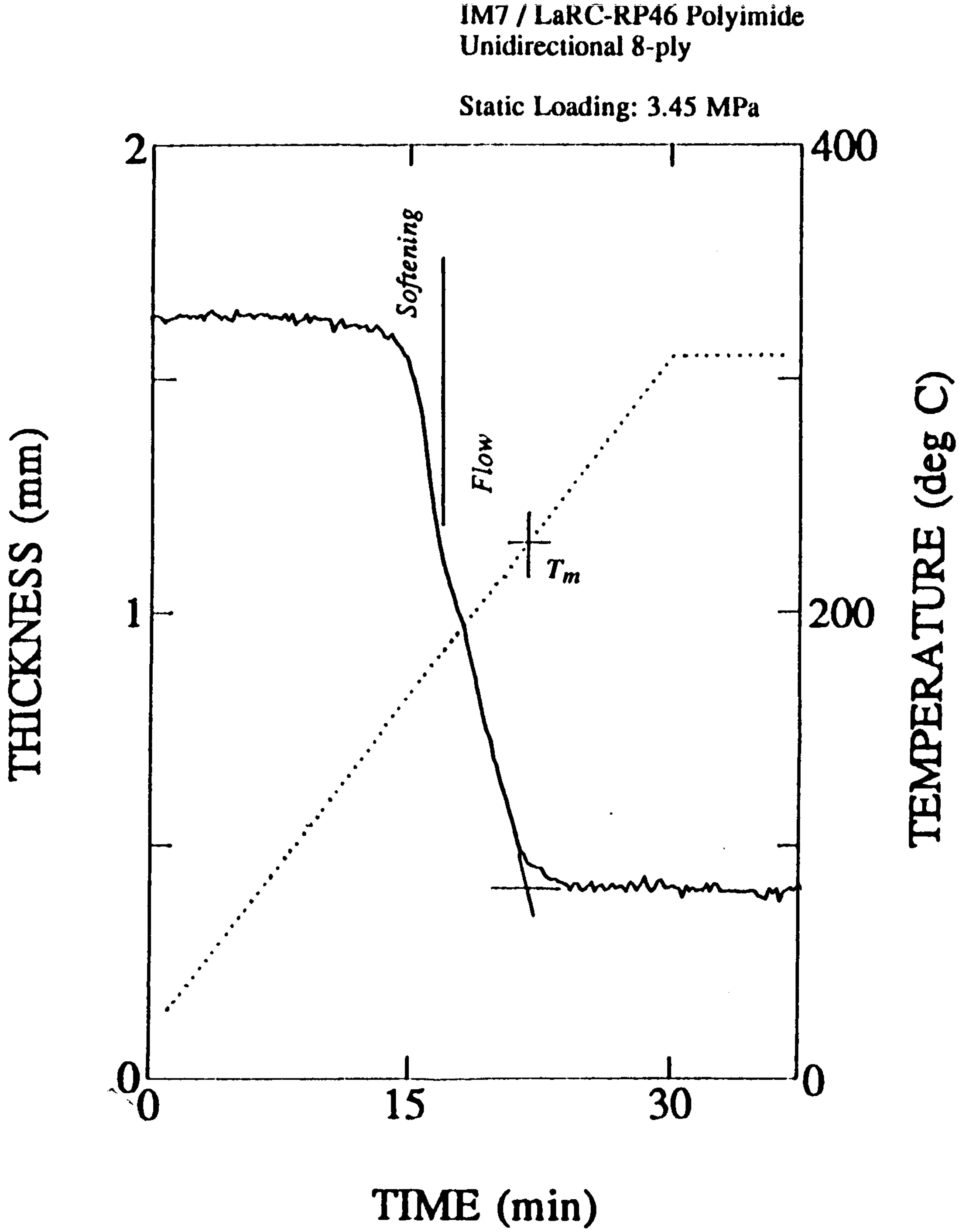
(3) In the plastometer curve, the initial half of the region with sudden thickness drop is caused by the softening of materials. Morphology study showed no flow in that region. The resin flow occurs in the second half of the thickness drop region. The melt transition temperature can be defined by extrapolating the drop region and the plateau of final thickness (Figure 1).

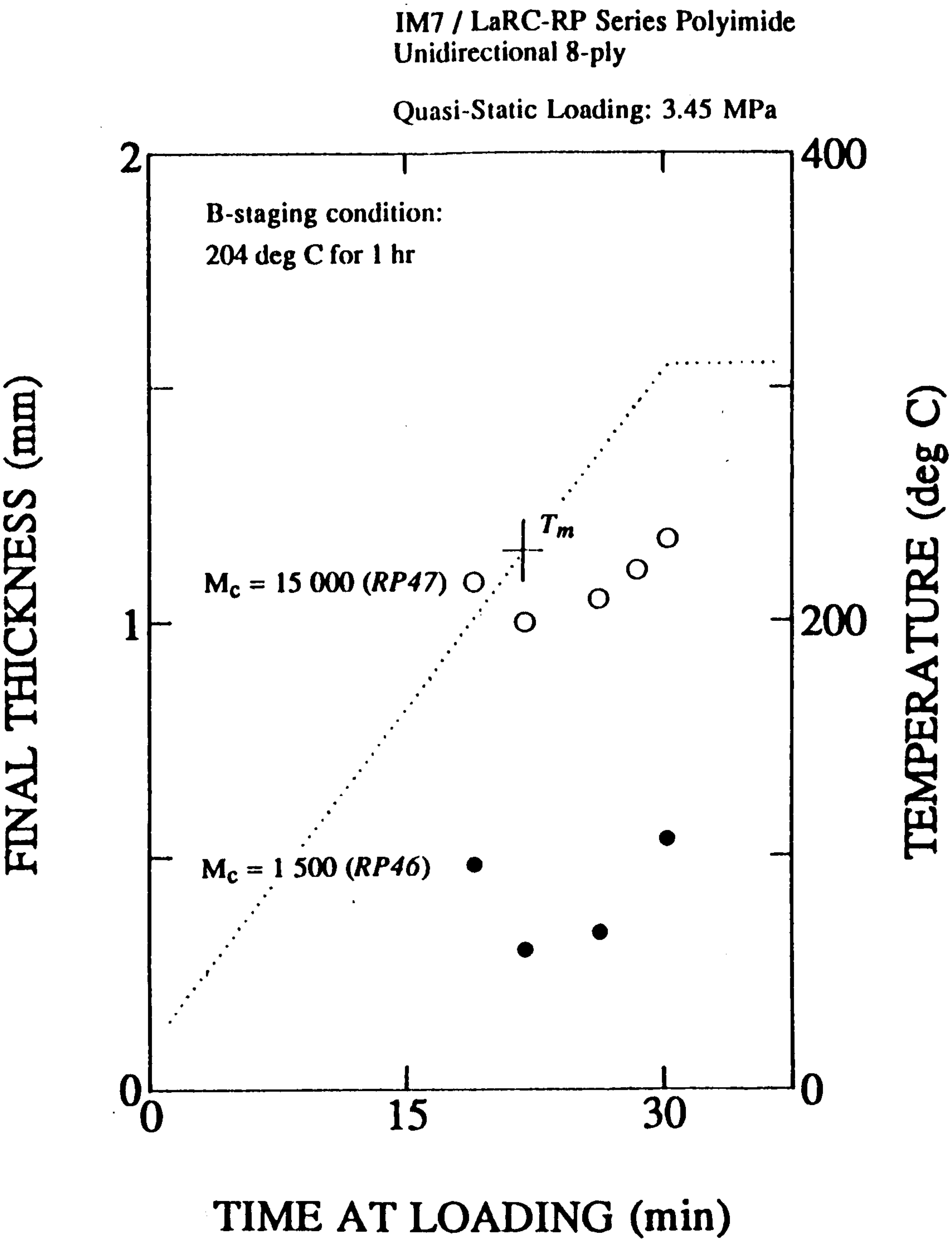
(4) A separate study indicated that the application of pressure at the melt transition temperature maximizes the squeeze flow (Figure 2). When the pressure is applied in the plateau region (Figures 1 and 2), a lesser amount of squeeze flow is observed due to the onset of crosslinking.

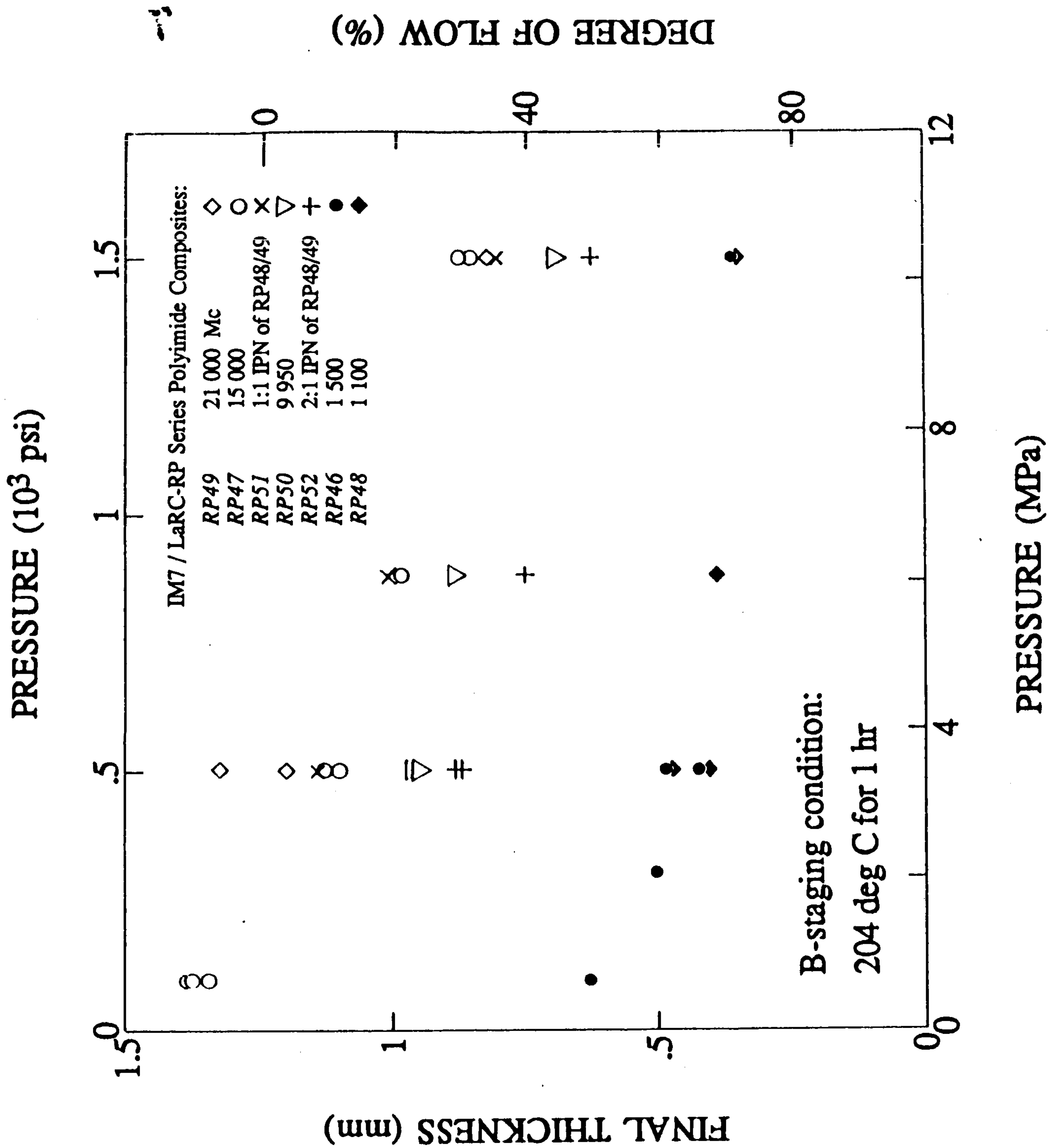
(5) The melt transition temperature, which constitutes the lower bound of processing window, is independent of Mc of resins but highly dependent on the degree of imidization. The upper bound of processing window is determined by the occurrence of gelation.

(6) Slower heating in cure cycle lowers the melt transition temperature and lengthens the gelation time of resin, thus widening the processing window.

(7) The increase of Mc of resin reduces the degree of matrix squeeze flow (Figure 3). The reduction of matrix resin flow can be counterbalanced by the application of higher pressure or the use of prepreg systems with higher volatile contents. The prepreg systems with higher volatile contents are prepared by B-staging at lower temperature.


IM7 / LaRC-RP46 Polyimide
Unidirectional 8-ply
Static Loading: 3.45 MPa
Softening
Flow
T_m
THICKNESS (mm)
TEMPERATURE (deg C)
TIME (min)
2
1
0
400
200
0
0
15
30


IM7 / LaRC-RP Series Polyimide
Unidirectional 8-ply
Quasi-Static Loading: 3.45 MPa
B-staging condition:
204 deg C for 1 hr
T_m
M_c = 15 000 (*RP47*)
M_c = 1 500 (*RP46*)
FINAL THICKNESS (mm)
TEMPERATURE (deg C)
TIME AT LOADING (min)
0
1
2
0
15
30
0
200
400



PRESSURE (10^3 psi)
DEGREE OF FLOW (%)
FINAL THICKNESS (mm)
PRESSURE (MPa)
IM7 / LaRC-RP Series Polyimide Composites:
RP49 21 000 Mc
RP47 15 000
RP51 1:1 IPN of RP48/49
RP50 9 950
RP52 2:1 IPN of RP48/49
RP46 1 500
RP48 1 100
B-staging condition:
204 deg C for 1 hr